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(54) Title: FLUORORUBBER COATING COMPOSITION

(57) Abstract

The invention provides a fluor elastomer coating composition comprising an organic solvent solution of fluor elastomer gum, a Schiff base which is the reaction product of an aminosilane compound, and preferably a partially hydrolyzed and polymerized aminosilane compound, with an aldehyde or ketone.

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FLUORORUBBER COATING COMPOSITION

5

This invention relates to a liquid coating composition containing fluoroelastomer. This composition is useful as a for coating for metallic gaskets used in automobiles.

- 10 Gaskets, such as cylinder gaskets for automobiles, are prepared by coating metallic substrates, such as stainless steel substrates, with a fluoroelastomer coating. Such gaskets are increasingly popular over gaskets made of asbestos. Japanese Unexamined Patent
15 Publication No. 2248453 discloses a fluoroelastomer coating composition comprising a polyol, a quaternary ammonium or phosphonium salt vulcanization accelerator, an amine vulcanization agent, a solvent mixture of ketone and alcohol and a magnesium oxide acid acceptor.
20 However, this composition exhibits poor adhesivity to substrates, and, as a result, requires the surface of the substrate to be coated with a primer before applying the fluoroelastomer coating composition. The use of the primer coating increases the time required
25 to produce a coated article, complicates the preparation process and increases the cost of the coated article.

- Japanese Examined Patent Publication No. 47-18,346 discloses a fluoroelastomer coating composition
30 containing an aminosilane compound. The aminosilane is supposed to improve bonding between the coating and substrates, however, the composition tends to gel prior to use, and once applied to a substrate, the coating has low durability against the long-life coolants (LLC)
35 and does not possess sufficient high initial adhesivity between the coating and the substrate--although

enhanced bonding between the coating and the substrate is acquired after aging the coated substrate.

U.S. Patent No. 4,960,918 discloses an organic silicon compound which is the condensation product of
5 α , w-bisaminopropylsiloxane and a cinnamaldehyde represented by the general formula PhCH=CHCHO . The compound is described as being a fluororubbers vulcanizing agent. However, such compounds are not believed to improve bonding between cured
10 fluoroelastomer coatings and metallic surfaces.

U.S. Patent No. 2,942,019 discloses the reaction product between an aldehyde or ketone and an organosilicon compound containing an aminoalkylsilyl group, such as certain aminopolysiloxanes. The patent
15 states that such compounds have various uses, including, for example, the use of trifunctional methylideneaminoalkylsiloxanes and substituted methylideneaminoalkylsiloxanes as thermosetting resins or as modifiers to thermosetting resins to improve
20 their color stability. The patent further states that the methylideneaminoalkylalkoxysilanes may be mixed or equilibrated with silicone elastomers before curing to improve their properties (column 7, lines 5-21). However, the patent does not disclose that the
25 compounds are useful as fluoroelastomer cure agents or that such compounds can enhance bonding between cured fluoroelastomers and metallic surfaces.

An urethane resin composition containing the reaction product of an amino silane compound and an
30 aldehyde or ketone compound and an isocyanate compound is disclosed in German Patent Application No. DE34148774. The application does not disclose that the reaction product of an aminosilane and aldehyde or ketone is useful as a latent fluoroelastomer
35 vulcanizing agent or bonding agent.

In one aspect, the invention provides a fluoroelastomer coating composition comprising an organic solvent solution of fluoroelastomer gum and a Schiff base. For the purposes of this invention, a
5 "Schiff base" is the reaction product of an aminosilane compound, or preferably a partially hydrolyzed and polymerized aminosilane compound hereinafter referred to as "polyaminosiloxane compounds", with aldehydes or ketones.

10 The proportions of components used in the compositions are set forth herein in parts by weight. Unless otherwise indicated, all amounts referred to are in parts by weight of the component per 100 parts by weight of the fluoroelastomer. For brevity, these
15 units are abbreviated as "phr".

In a preferred embodiment, the fluoroelastomer coating composition comprises, per each 100 parts by weight of fluoroelastomer gum, 2 to 50 phr by weight of the Schiff base, 1 to 30 phr acid acceptor.

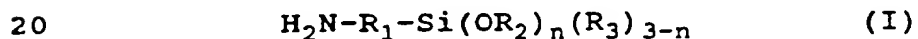
20 Most preferably, the coating composition further comprises other components commonly used to cure fluoroelastomers such as polyamines, polyols and peroxide vulcanization agents and those vulcanization accelerators (e.g., organoonium accelerators such as
25 quaternary ammonium and phosphonium salts), co-agents and acid acceptors commonly employed with the aforementioned vulcanization agents.

The present invention also provides a method of making a fluoroelastomer coated article comprising the
30 steps of: (A) applying the aforementioned fluoroelastomer coating composition to the inorganic (e.g., metal, glass or ceramic) surface of a substrate and (B) then heating the coated surface to 100 to 300°C, or, alternatively, irradiating the coated
35 surface with ionizing radiation at a level of 0.1 to 20

Mrads until the desired level of cure of the coating is effected.

The fluoroelastomer coating composition of this invention possesses good shelf-stability and does not
5 prematurely gel because the Schiff base will not react with the fluoroelastomer until the coated article is heated to a temperature adequate or irradiated sufficiently to decompose the Schiff base and liberate the aminosilane or polyaminosiloxane compounds.

10 Any aminosilane or polyaminosiloxane compound capable of enhancing the bonding between a fluoroelastomer and an inorganic surface, such as a metallic surface, is useful in the preparation of the Schiff base used in this invention. Many such
15 compounds are known in the art. Typically, such compounds contain at least one amino group, and aminosilane compounds may be represented by the following formula:



Where, in the above formula,

R_1 can be selected from the group consisting of $-\text{R}_4-$ and $-\text{R}_5-\text{NH}-\text{R}_6$, wherein R_4 , R_5 , and R_6 can be
25 independently selected from the group consisting of linear and branched alkylene groups, preferably having 1 to 4 carbon atoms, and most preferably having 3 carbon atoms, wherein among the aforementioned alkylene groups, propylene is particularly preferred; R_2 and R_3
30 can be independently selected from the group consisting of hydrogen atoms and lower alkyl groups having 1 to 5 carbon atoms (e.g., methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, and tert-butyl groups), and preferably 2 carbon atoms; and n is an integer of 2 to
35 3.

Representative examples of the aminosilane compounds useful in this invention include:

- 3-aminopropyltrimethoxysilane,
- 3-aminopropyltriethoxysilane,
- 5 3-aminoisobutyltrimethoxysilane,
- 3-aminoisobutyltriethoxysilane,
- N-2-aminoethyl-3-aminopropyltrimethoxysilane,
- N-2-aminoethyl-3-aminopropyltriethoxysilane,
- 3-aminopropyldimethoxymethylsilane,
- 10 3-aminopropyldiethoxymethylsilane,
- 3-aminopropyldimethoxyethylsilane, and
- 3-aminopropyldiethoxyethylsilane.

Preferably, the alkoxy groups of the aminosilane compound are partially hydrolyzed and the aminosilane
15 compound is partially polymerized by heating the aminosilane with an equimolar amount of water at a temperature between 40 and 100°C to yield a polyaminosiloxane product. To monitor the progress of the aforementioned reactions, gel permeation
20 chromatography (to monitor the molecular weight of the reaction mixture) or infrared spectrophotometry (to measure the increasing concentration of hydroxyl groups in the reaction mixture) can be used. A more practical means of monitoring the progress of the reaction is
25 simply to measure the viscosity of the reaction mixture. Typically, the initial viscosity of the reaction mixture is about 1 to 2 cps at 25°C, and after reaction, the reaction mixture will possess a viscosity of 3 to 10,000 cps, and preferably 5 and 1000 cps, at
30 25°C.

To the polyaminosiloxane, additional tetraalkoxysilane compounds may be added. The tetraalkoxysilane compounds may be reacted with water as described above prior to their addition to the
35 polyaminosiloxane, or they may be hydrolyzed along with the aminosilane compound. Preferably, the alkyl group

of the alkoxy moiety of the tetraalkoxysilane compound is a lower alkyl such as methyl or ethyl. The tetraalkoxysilane compounds may be added to the coating composition in ratios of 1:100 to 2:1 parts by weight

5 tetraalkoxysilane compound to aminosilane or polyaminosiloxane compound. If a ratio greater than 2:1 used, the shelf life of the coating composition may be shortened. If a ratio smaller than 1:100 is used, the adhesion to the substrate will be poor.

10 The aldehyde or ketone compounds useful in the preparation of the Schiff base can be represented by the following formula:

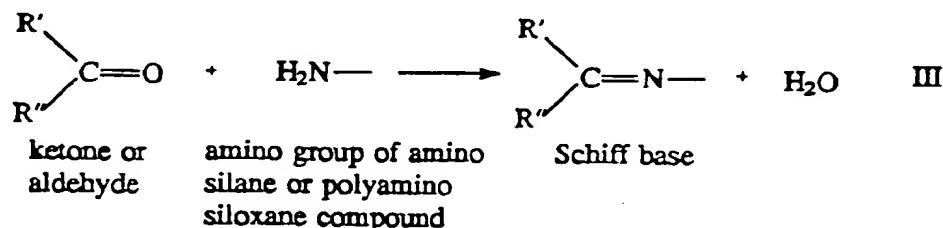


Where, in the above formula, R_4 and R_5 can be independently selected from the group consisting of hydrogen atoms, and linear or branched alkyl groups having 1 to 12 carbon atoms, aryl groups, and alkaryl groups (wherein the alkyl moiety that is part of the alkaryl group is a lower alkyl moiety having 1 to 5 carbon atoms, e.g., methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, tert-butyl, and linear or branched pentyl moieties), or R_4 and R_5 may together, with the carbon to which they are both bonded, form an unsubstituted or substituted alkylene group having 3 to 10, and preferably 6, carbon atoms, in which case the ketone will be a cycloalkanone and the aldehyde a cinnamaldehyde.

Representative examples of ketone or aldehyde compounds useful in this invention include methylethyl ketone, acetone, cyclohexanone, methylbutyl ketone, methylisobutyl ketone, acetophenone,

ethylcyclohexanone, formaldehyde, acetaldehyde, propylaldehyde, butylaldehyde, and cinnamaldehyde.

The Schiff base is prepared by reacting the aminosilane or polyaminosiloxane compound with the
 5 desired aldehyde or ketone. In the reaction, the amino group of an aminosilane or polyaminosiloxane compound and the oxygen of the aldehyde or ketone are believed to undergo the following dehydration and condensation reaction:



10 The reaction depicted above is accomplished by heating the reactants to 50 to 150°C for a period of 10 to 120 minutes in the presence of an entrainer capable of forming an azeotrope with water (e.g., cyclohexane or toluene). Thereby forming the Schiff base and
 15 removing the water from the reaction product. The reaction may be accelerated by use of a catalyst such as an acid catalyst (e.g., toluenesulfonic acid or benzenesulfonic acid).

Typically, the amount of the Schiff base added to
 20 the coating composition is 2 to 50 phr. When less than 2 phr of Schiff base is used in the composition, the coating is inadequately cross-linked and will not possess desirable physical properties, have adequate heat resistance or be adequately bonded to the
 25 substrate surface. If more than 50 phr of the Schiff base are used in the coating composition, the cured coating may be too hard.

Fluoroelastomers useful in the coating composition include any fluoroelastomers known in the art. Some of

commercially available fluoroelastomers useful in the coating composition include copolymers of vinylidene fluoride and hexafluoropropylene, terpolymers of vinylidene fluoride, hexafluoropropylene and
5 tetrafluoroethylene, copolymers of tetrafluoroethylene and propylene, and terpolymers of vinylidene fluoride, tetrafluoroethylene and propylene.

The coating compositions of the invention may further comprise acid acceptors and conventional
10 vulcanizing agents and vulcanization accelerators known in the art (e.g., polyamine compounds, polyols, peroxide compounds and their conventional coagents or cocuratives, and organoonium vulcanization accelerators). The additional curatives permit the
15 formulator to enhance the cure rate of the coating composition.

When an additional vulcanizing agent is employed in the coating composition, the rate of vulcanization can be controlled and accelerated more easily than when
20 the Schiff base is used as the sole vulcanizing agent in the coating composition. Though the amount of the additional vulcanizing agent added to the coating composition may be varied depending upon the particular physical properties desired in the cured
25 fluoroelastomer coating, generally, 0.5 to 10 phr of polyamine and polyol vulcanizing agent are employed because such levels provide good cure rates and coating composition shelf life. Generally, amounts of 0.1 to 10 phr of peroxide vulcanizing agent and 1 to 20 phr of
30 cocurative or coagent are employed.

Representative examples of the polyamine vulcanizing agents include N,N'-dicinnamylidene-1,6-hexanediamine, hexamethylenediamine monocarbonate, and
4,4'-methylenebis(cyclohexylamine) carbamate.

35

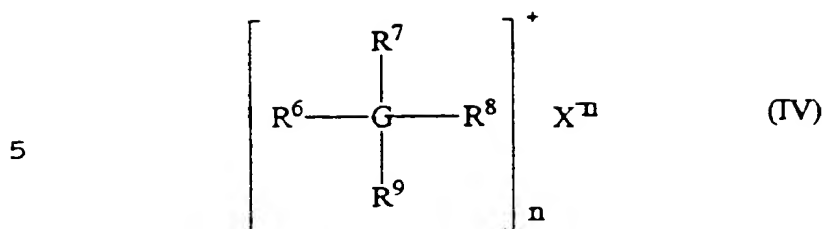
Representative examples of polyols include 2,2-bis(4-hydroxyphenyl)propane, 2,2-bis(4-hydroxyphenyl)perfluoropropane, hydroquinone, catechol, resorcin, 4,4-dihydroxyphenyl, 4,4-dihydroxydiphenyl
5 methane, 4,4-dihydroxydiphenyl sulfone, and 4,4-dihydroxydiphenyl mercaptan. The alkali metal salts and alkaline earth metal salts of such polyols are also useful.

Representative examples of peroxide vulcanization
10 agents include organic or inorganic peroxides such as benzoyl peroxide, bis(2,4 dichlorobenzoyl)peroxide, dicumyl peroxide, t-butylhydroperoxide, di-t-butyl peroxide, t-butylperoxy benzoate, and lauroyl peroxide, 2,5-dimethyl-2,5-bis(t-butylperoxy)hexane, 1,1-bis(t-
15 butyl)3,3,5-trimethylcyclohexane and 2,5-dimethyl-2,5-bis(t-butylperoxy)hexyne-3. Preferably, a cocuring agent is used in conjunction with the peroxide vulcanization agent. Examples of cocuring agents include triallylcyanurate, diallylphthalate,
20 allylmethacrylate and triallylisocyanurate.

The vulcanization accelerators mentioned above are known and include any capable of functioning in conjunction with vulcanizing agent such as a polyamine or polyol compound as a vulcanizing accelerator. For
25 example, the accelerators include organo-onium compounds such as the phosphonium, ammonium and sulfonium compounds described in U.S. Patent Nos. 4,882,390, 4,233,421 and 5,086,123.

Typical phosphonium compounds include, for
30 example, amino-phosphonium, phosphorane, and phosphorous-containing-iminium compounds. Some of the phosphonium and ammonium compounds particularly useful in this invention can be represented by the following general formula:

35



where, in the above formula, G is a phosphorous or
 10 nitrogen atom, X^n is an organic or inorganic anion
 such as halide, hydroxylate, alkoxylate, carboxylate,
 phenoxide, sulfonate, sulfate, sulfite, carbonate and
 nitrate, n is equal to the valence of the anion X and
 R^6 , R^7 , R^8 and R^9 are selected from the group of radicals
 15 consisting of alkyl, aryl, alkenyl or combinations
 thereof. R^6 , R^7 , R^8 and R^9 can be substituted with
 chlorine, fluorine, bromine, cyano, $-\text{OR}^{16}$ and $-\text{COOR}^{16}$
 moieties where R^{16} is selected from the group consisting
 of C_1 to C_{20} alkyl, aryl, aralkyl and alkenyl. Any pair
 20 of said R^{6-9} groups can be connected with each other and
 the nitrogen or phosphorus atom to form a heterocyclic
 ring.

Representative examples of useful organoonium
 compounds include tetraphenyl phosphonium chloride,
 25 tetraphenyl phosphonium bromide, tetraoctyl phosphonium
 chloride, tetra-n-butyl phosphonium chloride,
 tetraethyl phosphonium chloride, tetramethyl
 phosphonium chloride, tetramethyl phosphonium bromide,
 triphenylbenzyl phosphonium chloride, triphenylbenzyl
 30 phosphonium bromide, triphenylbenzyl phosphonium
 stearate, triphenylbenzyl phosphonium benzoate,
 triphenylisobutyl phosphonium bromide, trioctyl-n-butyl
 phosphonium chloride, trioctylbenzyl phosphonium
 chloride, trioctylbenzyl phosphonium acetate,
 35 triphenyl-2,4-dichlorobenzyl phosphonium chloride,
 trioctylmethoxyethoxyethyl phosphonium chloride,

triphenylethoxycarbonylmethyl phosphonium chloride,
triphenylallyl phosphonium chloride, 1-alkylpyridinium
salts, 5-alkyl-1,5-diaza-bicyclo[4,3,0]-5-nonenium
salts, 8-alkyl-1,8-diazabicyclo-[5,4,0]-7-undecenium
5 salts, tetraphenyl ammonium chloride, tetraphenyl
ammonium bromide, tetraoctyl ammonium chloride, tetra-
n-butyl ammonium chloride, tetraethyl ammonium
chloride, tetramethyl ammonium chloride, tetramethyl
ammonium bromide, triphenylbenzyl ammonium chloride,
10 triphenylbenzyl ammonium bromide, triphenylbenzyl
ammonium stearate, triphenylbenzyl ammonium benzoate,
triphenyl-isobutyl ammonium bromide, trioctyl-n-butyl
ammonium chloride, trioctylbenzyl ammonium chloride,
trioctylbenzyl ammonium acetate, triphenyl-2,4-
15 dichlorobenzyl ammonium chloride, trioctyl
methoxyethoxyethyl ammonium chloride, triphenyl
ethoxycarbonyl methyl ammonium chloride, and triphenyl
allyl ammonium chloride.

Preferably, vulcanization accelerators are used in
20 in composition in amounts of 0.5 to 10 phr.

Acid acceptors useful in the invention can be any
of the those compounds which are generally used in the
vulcanization of fluoroelastomers. Examples of such
compounds include divalent metal oxides and hydroxides,
25 and they may be employed in the coating composition
either singly or in mixtures of two or more acid
acceptors. Representative examples of acid acceptors
include magnesium oxide, calcium oxide, zinc oxide,
lead oxide, and calcium hydroxide. Among
30 aforementioned acid acceptors, magnesium oxide is
particularly preferably because of its low activity.
Typically, acid acceptor is added to the composition at
a level of from 1 to 30 phr. If less than 1 phr or
acid acceptor is used, vulcanization of the
35 fluoroelastomer is slow, and if more than 30 phr of

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acid acceptor is used, the cured fluoroelastomer coating has poor water resistance.

- The fluoroelastomer coating composition of the present invention can be produced by mixing the various components mentioned above with an organic solvent capable of dissolving the components of the mixture. Useful organic solvents include those selected from the group consisting of methyl isobutyl ketone, methylethyl ketone, ethyl acetate, cellosolve acetate, methanol, ethanol and isopropanol, and the solvents can be used singly or in mixtures of two or more. The organic solvent is used in such an amount that the resultant solution has a total solid content of from 10 to 90% by weight based on the total weight of the composition.
- The exact total solid content of the coating will depend on the coating method selected to apply the composition. This range is preferred because lower solids solutions will possess unduly low viscosities and cannot easily be applied to a metallic plate or the like and the resultant coating requires a long drying time. If the solid content is greater than 90%, the coating solution will possess an undesirably high viscosity and be difficult to uniformly apply to substrates.
- Additional adjuvants and fillers commonly used in fluoroelastomer compositions may also be added to the coating composition. For example, fillers such as, for example, silica, clay, diatomaceous earth, talc, carbon black, carbon fibers, alumina fibers, glass fibers, and asbestos fibers may be added. Although the amount of filler to be added will depend upon the particular use and physical properties contemplated, amounts in the range of 5 to 200 phr, or preferably of 10 to 100 phr, may be used.
- The coating composition of the present invention can be applied to a substrate surface by any

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conventional coating method, for example, spray coating, dip coating, flow coating and roll coating methods may be used. After coating the substrate, the coated substrate is heated to 100°C to 300°C, preferably 120°C to 250°C, and most preferably 120°C to 200°C, for about 15 minutes to 2 hours, and preferably for 30 to 60 minutes. The heating is believed to decompose the Schiff base and liberate the active aminosilane or aminopolysiloxane compound resulting in vulcanization of the coating and bonding of the coating to the substrate.

Examples 1-7 and Comparative Examples C1-C3

The following procedure was utilized to synthesize the Schiff bases used in the subsequent examples.

Preparation of an aminosilane Schiff base. A three-neck flask equipped with a reflux tube was charged with 221.14 g (1.0 mol) of 3-aminopropyltriethoxysilane and 500 g of toluene. Dry nitrogen was bubbled through the liquid to displace entrapped air. To the flask, 132.16 g (1.0 mol) of cinnamaldehyde was added dropwise and the resulting mixture was stirred at a room temperature for 30 minutes. The temperature of the contents of the flask was then increased to 100 to 120°C and held there for two hours with stirring to remove the toluene. The remaining contents of the flask was then vacuum distilled to remove remaining volatile matter and obtain N-cinnamylidene-3-aminopropyl triethoxysilane (hereinafter referred to as "CAPS").

Preparation and evaluation of the fluoroelastomer coating compositions. The exemplified fluoroelastomer compositions were prepared by compounding the following ingredients using a two-roll mill: 100 phr of

fluoroelastomer (commercially available as FLUOREL fluoroelastomer FC-2145 from 3M), 30 phr of carbon black (commercially available as Thermal N-900 carbon black from Cancarb Ltd.), magnesium oxide

- 5 (commercially available as "Kyowamag" #30 from Kyowa Kagaku K.K.) in the amount specified in Table 1, and N,N'-cinnamylidene-1,6-hexadamine in the amount specified in Table-1 (commercially available as "Diak" #3 from DuPont DeNemours and Company).

- 10 To each of the compounded fluoroelastomer compositions described above, a solution of either the CAPS (described above) or 3-aminopropyl triethoxy silane (hereinafter referred to as "APS") was added. The solution was prepared by dissolving the CAPS or
15 APS, in the amount specified in Table-1, in 300 g of methyl isobutyl ketone (MIBK) and then combining the resulting solution to the compounded fluoroelastomer compositions described above.

- The fluoroelastomer coating composition thus
20 obtained was applied using a bar coater to a stainless steel plate (SS-301) cleaned with 1,1,1-trichloroethylene. The coating was dried at a room temperature and then further dried at 120°C for 10 minutes to yield a coating of about 20 micrometers in
25 thickness. The coating was then vulcanized at 200°C for 30 minutes.

- To evaluate the adhesivity or bonding of the fluoroelastomer coating compositions, the coatings, immediately after the vulcanizing treatment and after
30 immersion for 500 hours in LLC (available commercially from Toyota Motor Company as Genuine Long-Life Coolant-diluted to 50% by volume with water) at 120°C were subjected to a "Cross-Cut Adhesion" test and a "Pencil Hardness" test in accordance with the procedures set
35 forth in Japanese Industrial Standard (JIS) K-5400. The results of this testing are presented in Table-1.

The stability of the fluoroelastomer coating compositions at normal room temperature were also evaluated. The number of days required for the coating compositions to gel are presented in Table-1.

- 5 In the "Pencil Hardness Test", a rating of "H" indicates the hardest possible coating, "HB" the next hardest coating, "B" the next hardest coating and "2B" the least hard coating. It is important to achieve coatings having an acceptable level of initial
- 10 hardness. The acceptable level of hardness will vary depending upon the application, and initial hardnesses of HB to 2B may be acceptable. Perhaps even more important than initial coating hardness is whether the coating hardness changes over time as the coating is
- 15 exposed to the environmental conditions normally encountered in use. Generally, large changes in the Pencil Hardness Test rating is undesirable.

- In the Cross-Cut Adhesion Test, the vulcanized coating is scored or cut at regular intervals of 1.0 mm
- 20 to form ten sections. A tape is then applied to the cut coating and removed. Any coating that does not adhere to the substrate is removed by the tape. The coating adhesion is then measured by counting the number of sections remaining bonded to the substrate.
- 25 A rating of 10 indicates that no coating was lost after tape removal. While any loss of coating is undesirable, some loss of the coating, may not render a composition unacceptable.

Table 1

	Example							Comparative Example		
	1	2	3	4	5	6	7	1	2	3
Fluoroelastomer	100	100	100	100	100	100	100	100	100	100
MT carbon	30	30	30	30	30	30	30	30	30	30
Magnesium oxide	10	10	10	10	5	20	10	10	10	-
Diak #3	0	0	0	0	0	0	3	0	3	0.1
CAPS	5	10	20	40	10	10	10	0	0	1.0
APS	0	0	0	0	0	0	0	10	0	0
MIBK	300	300	300	300	300	300	300	300	300	300
Initial Pencil Hardness Cross-cut Adhesion	HB 10	HB 10	HB 10	B 10	HB 10	B 10	B 10	H 10	H 10	B 0
After immersion Pencil Hardness Cross-cut Adhesion	HB 6	HB 10	HB 10	B 10	HB 10	B 10	HB 10	HB 8	2B 0	B 0
Gelation	>10	>10	>10	>10	>10	>10	>10	≤3	>10	>10

The data shows that the coatings of this invention have good pot life, good adhesion both before and after immersion in LLC and good hardness both before and
5 after immersion in LLC.

CLAIMS

1. A fluoroelastomer coating composition, comprising
5 for each 100 parts by weight of fluoroelastomer:
 (A). 2 to 50 parts by weight of a Schiff base
 prepared by reacting an aminosilane or
 polyaminosiloxane compound with an aldehyde or
 ketone compound; and
10 (B) 1 to 30 parts by weight of an acid acceptor;
and such an amount of an organic solvent as to give the
composition a total solid content of 10 to 90% based on
the total weight of the composition.
- 15 2. A fluoroelastomer coating composition according to
claim 1 wherein the composition further comprises an
amine vulcanizing agent.
3. A fluoroelastomer coating composition according to
20 claim 2 wherein the amine vulcanizing agent is present
in the composition in an amount of 0.5 to 10 parts by
weight per 100 parts by weight of the fluoroelastomer.
4. A fluoroelastomer coating composition according to
25 claim 2 wherein the composition further comprises an
organoonium vulcanization accelerator.
5. A fluoroelastomer coating composition according to
claim 1 wherein the composition further comprises
30 polyol and organoonium vulcanization accelerator.
6. A fluoroelastomer coating composition according to
claim 5 wherein the polyol is present in the
composition in an amount of 0.5 to 10 parts per 100
35 parts of fluoroelastomer and the organoonium compound

is present in the composition in an amount from 0.5 to 10 parts per 100 parts of fluoroelastomer.

7. A fluoroelastomer coating composition according to claim 1 wherein the composition further comprises 0.1 to 10 parts by weight of peroxide vulcanizing agent per 100 parts by weight of fluoroelastomer and 1 to 20 parts by weight of coagent per 100 parts by weight of fluoroelastomer.

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7. An article coated with the fluoroelastomer composition of claim 1.

8. A method of producing a coated article comprising the steps of:

(A) applying a coating of the fluoroelastomer coating composition of claim 1 to the surface of an inorganic substrate; and
(B) then heating the resultant coated article to a temperature in the range of from 100 to 300°C until the desired degree of cure of the coating is achieved.

10. A method of producing a coated article comprising the steps of:

(A) applying a coating of the fluoroelastomer composition of claim 1 to the surface of an inorganic substrate; and
(B) then irradiating the resultant coated substrate with radiation of sufficient intensity to effect a cure of the fluoroelastomer coating.

INTERNATIONAL SEARCH REPORT

International Application No
PCT/US 93/11420

A. CLASSIFICATION OF SUBJECT MATTER

IPC 5 C09D127/12 C08K13/00 B05D1/00 //(C08K13/00,5:54,3:00,
5:00)

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 5 C08L C08K C09D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US,A,3 656 530 (EVANS, MECK, THIBODEAU) 18 April 1972 see claim 1	1
A	US,A,4 960 918 (KONNO, YAMAMOTO, SONEGAWA) 2 October 1990 cited in the application see column 2, line 54 - line 60; examples 5-6	1,8

☐ Further documents are listed in the continuation of box C.☒ Patent family members are listed in annex.

* Special categories of cited documents:

- *A* document defining the general state of the art which is not considered to be of particular relevance
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- *L* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- *O* document referring to an oral disclosure, use, exhibition or other means
- *P* document published prior to the international filing date but later than the priority date claimed

- *T* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
- *X* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
- *Y* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
- *&* document member of the same patent family

Date of the actual completion of the international search

5 April 1994

Date of mailing of the international search report

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/US 93/11420

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US-A-3656530	18-04-72	NONE	
US-A-4960918	02-10-90	JP-C- 1755666	23-04-93
		JP-A- 2088585	28-03-90
		JP-B- 4038758	25-06-92

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